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(54) MANUFACTURING PROCESS FOR MIXED NUCLEAR FUEL PELLETS

(71) We, BELGONUCLEAIRE S.A., a Belgian Body Corporate, of rue des Colonies 35, B-1000, Brussels, Belgium, do hereby declare the invention, for which we pray that a patent may be granted to us and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to a process for producing nuclear reactor fuel, and particularly compressed mixed fuel comprising fertile and fissile material.

Such compressed fuel, generally manufactured from ceramic powders such as, for example, oxides, carbides or nitrides or uranium, plutonium or other transuranian elements, will be hereinafter designated by the word "pellets". Ceramic pellets are generally prepared according to a method including the steps of pelletizing a raw material, sintering the raw pellets and machining the sintered pellets in order to comply with the very strict specifications set by clients.

In the case of mixed ceramic pellets, these steps are preceded by a proportioning and mixing of the raw powders. All parameters connected with these steps must be scrupeously observed in order to manufacture pellets in a reproducible way. Indeed, if one of the characteristics of the raw powder or of the mixing, sintering or pelletizing conditions changes slightly during the production of a batch of pellets, the products obtained will not be uniform.

During the manufacture of a batch of pellets, some pellets are however rejected because of flaws, and should preferably be recycled. This need for recycling is more evident, when the mixed ceramic pellets comprise fertile and fissile material, because otherwise the fissile material would have to be stored. This scrap, i.e. the rejected pellets, may easily be recycled after being submitted to an appropriate mechanical and/or thermal treatment, transforming them into a powder which may be added to the raw powder. If, however, the characteristics of the recycled powder are different from those of the raw powder, all other parameters and characteristics remaining the same, the percentage in weight of recycled powder to be added to the raw powder should be well defined and constant, in order to obtain sintered pellets with reproducible characteristics.

If the characteristics of the recycled powder are different from those of the raw powder, during the production of a batch of pellets of a certain type, the following procedures may be adopted:

1) prepare the pellets of a batch starting from fresh powder only and recycle the scrap after the fabrication cycle by modifying one or several fabrication parameters. This process, however, implies a supplementary adjustment, as well as the storage of fissile material;
2) systematically break up, at the beginning of an operational process a sufficient quantity of sintered pellets in order to have available, from the initial production, an amount of recycled powder which enables the adoption of one and the same fabrication technique without modifying the parameters. This technique is, however, expensive and should be avoided when the pellets contain fissile material.

It is an object of this invention to provide a manufacturing process for mixed ceramic pellets which enables the provision, during the fabrication of a batch of pellets, of the same percentage by weight of recycle powder in the fresh powder without for this purpose breaking down pellets at the beginning of the cycle.

According to the invention in a process for producing mixed ceramic pellets consisting of fertile and fissile material, including the steps of proportioning the constituent powders, mixing said powders, pelletizing the mixture and sintering the pellets thus obtained, the fabrication scrap is recycled and added to the constituent powders, and

the percentage (by weight) of recycled powder in the constituent powders is maintained constant by substituting for the recycled powder derived from the fabrication scrap, both at the beginning of the fabrication procedure and during said procedure, when there is a lack of recycled powder derived from fabrication scrap, an amount of recycled powder derived from a similar fabrication of pellets consisting only of fertile material.

During the fabrication cycle, the fertile material will be gradually replaced by mixed recycled powder in proportion to the available quantities of scrap, without, however, exceeding the total percentage of recycled powder predetermined at the beginning of the fabrication procedure. In order to have available, during the whole fabrication procedure, recycled powder having the same characteristics, recycled powder derived from the fertile constituent only is prepared in advance by a process which is the same as for the fabrication cycle.

This method thus allows the fabrication scrap to be recycled as it is obtained, no matter how much there may be, without being obliged to store scrap containing fissile material or to break down good pellets. This process moreover, allows a complete batch of pellets to be manufactured without scrap and without having to modify the parameters during the procedure.

The invention will hereinafter be described in more detail with reference to a non-limitative example. Before starting a fabrication process of a 4 ton batch of pellets with a density of 93% of the theoretical density and comprising 96% uranium oxide and 4% plutonium oxide, 50 kg uranium oxide pellets of the same density are prepared.

Indeed, on a 4 ton production, the total amount of rejected pellets may be estimated at 5%, i.e. 200 kg. In order to recycle this 200 kg of scrap according to the process of the invention, within the fabrication procedure as it is formed, about 50 kg scrap comprising only uranium oxide will be needed.

a) Fabrication of uranium oxide scrap.

Uranium oxide powder with a specific surface of 3 m^2/g, an apparent density of 2 g/cm^3, and a granulometry below 100 micron, is compressed with the help of a mechanical press, at a pressure of 3 t/cm^2 into pellets with a green density of 5.5 g/cm^3. These pellets are then sintered for four hours in an oven at a temperature of 1650° C and in an argon atmosphere containing 5% hydrogen. The pellets thus obtained have a density of 93% of the theoretical density. 50 kg of these pellets are ground, first in a hammer-mill and afterwards in a conical ball mill, in order to obtain a powder with a granulometry of less than 80 micron, a specific surface of 1 m^2/g and an apparent density near to 2.5 g/cm^3.

b) Fabrication of UO_2—PuO_2 pellets.

These pellets are manufactured starting from:

—UO_2 powder with a specific surface of 3 m^2/g, an apparent density of 2 g/cm^3, and a granulometry of less than 100 micron,

—PuO_2 powder with a specific surface of 8 m^2/g, an apparent density of 2 g/cm^3, and a granulometry below 50 micron,

—recycled powder.

In order not to be obliged to change the parameters during the process, the fabrication procedure starts with the following proportions of the constituent powders:

—5% (by weight) of recycled UO_2 powder manufactured according to a) above;

—91% by weight of fresh UO_2 powder;

—4% by weight of fresh PuO_2 powder.

These constituents are mixed and homogenized in a screw-mixer. The mixed powder is then compressed at 3 t/cm^2 in a mechanical press into pellets with a green density of 5.5 g/cm^3. The obtained pellets are sintered for four hours in an oven at 1650° C, under argon containing 5% hydrogen. These pellets which will then have a density of 93% of the theoretical density, are subjected to severe control.

The pellets rejected for surface flaws or for non-conformity with the specified diameter or density, are ground as they appear, with the same equipment used for the grinding of the UO_2 pellets according to a) above (hammer-mill and conical ball mill), in order to obtain a fine powder containing 96% UO_2 and 4% PuO_2. The characteristics of this powder are almost identical with those of the recycled pure UO_2 powder (granulometry below 80 microns, specific surface 1 m^2/g, apparent density 2.5 g/cm^3).

The recycled powder is added in proportion as it is obtained to the mixture of fresh UO_2 and PuO_2 powder, as a substitute for the recycled UO_2 and in a maximum amount of 5%.

During fabrication, after the production of 100 kg, the initial mixture is modified in the following way:

—2% in weight of recycled UO_2 powder;

—91.12% in weight of fresh UO_2 powder;

—3.88% in weight of fresh PuO_2 powder;

—3% in weight of recycle UO_2—PuO_2 powder.

If, during the production, the quantity of scrap increases, the proportioning of the mixture may be modified by adding an amount of 5% recycled UO_2—PuO_2 powder. The mixture will then consist of:

—91.2% in weight of fresh UO_2 powder;

—3.8% in weight of fresh PuO powder;

—5% in weight of recycled UO_2—PuO_2 powder.

Thus the 4 ton pellets have been manufactured without changing the parameters influencing the fabrication, and with a constant quantity of recycled powder.

The process of the present invention described above is in no way limitative and obviously modifications may be incorporated within the scope of the invention.

WHAT WE CLAIM IS:—

1. A process for producing mixed ceramic pellets consisting of fertile and fissile material including the steps of proportioning the constituent powders, mixing said powders, pelletizing the mixture and sintering the pellets thus obtained, wherein fabrication scrap is recycled and added to the constituent powders and wherein the percentage by weight of recycled powder in the constituent powders is maintained constant by substituting for the recycled powder derived from the fabrication scrap both at the beginning of the fabrication procedure and during said procedure when there is a lack of recycled powder derived from fabrication scrap, an amount of recycled powder derived from a similar fabrication of pellets consisting only of fertile material.

2. A process for producing mixed ceramic pellets according to claim 1, wherein the recycled powder derived from the fabrication scrap is added to the constituent powders as it is obtained.

3. A process for producing mixed ceramic pellets, according to claim 1, wherein the production starts by adding to the constituent powders a predetermined percentage of recycled powder derived from pellets, consisting only of fertile material, and wherein, as soon as fabrication scrap is obtained, the recycled powder from the pellets comprising only fertile material is replaced at least in part by powder derived from the fabrication scrap.

4. A process for producing mixed ceramic pellets according to any of the preceding claims, wherein the pellets comprise a mixture of uranium oxide and plutonium oxide.

5. A process for producing mixed ceramic pellets consisting of fertile and fissile material substantially as hereinbefore described.

6. Ceramic pellets consisting of a mixture of fertile and fissile material produced according to the process claimed in any one of the preceding claims.

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РОССИЙСКОЕ АГЕНТСТВО
ПО ПАТЕНТАМ И ТОВАРНЫМ ЗНАКАМ

(12) ОПИСАНИЕ ИЗОБРЕТЕНИЯ К ПАТЕНТУ РОССИЙСКОЙ ФЕДЕРАЦИИ

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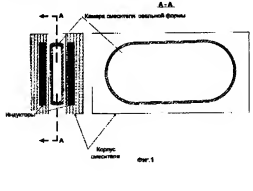
(54) СПОСОБ ПОЛУЧЕНИЯ ГОМОГЕННОГО ЯДЕРНОГО ТОПЛИВА

(57)

Изобретение относится к области атомной техники и может быть использовано для получения гомогенного ядерного топлива из смеси оксидов урана и плутония. Способ включает загрузку в камеру порошков оксидов урана и плутония и магнитных игл, смешивание порошков оксидов урана и плутония с помощью магнитных игл, прессование полученной смеси порошков в таблетку и спекание таблеток. Магнитные иглы перемещаются в камере под воздействием переменного магнитного поля. Камеру заполняют на 70 - 90% ее объема порошками оксидов урана, плутония и магнитными иглами. При этом отношение суммарной массы порошков оксидов урана и плутония к массе магнитных игл задают от 0,30 до 0,65, преимущественно от 0,40 до 0,50. Затем камеру вместе с порошками и магнитными иглами подвергают глубокому охлаждению и проводят смешивание порошков. Технический результат: повышение дисперсности, равномерности перемешивания, насыпной массы и текучести пресс-порошка. Улучшение этих характеристик повышает производительность процесса и обеспечивает ядерную безопасность при смешивании. 6 з.п. ф-лы, 2 ил., 1 табл.



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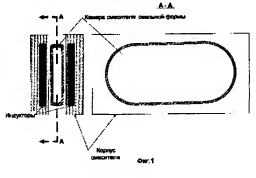
(54) METHOD FOR PRODUCING HOMOGENEOUS NUCLEAR FUEL

(57) Abstract:

FIELD: atomic engineering; nuclear fuel production from mixture of uranium and plutonium oxides. SUBSTANCE: method includes charging the chamber with uranium and plutonium oxide powders and magnetic needles, mixing up these powders by means of magnetic needles, compressing mixture obtained to form pellets, and sintering the pellets. Magnetic needles are moving within chamber under action of variable magnetic field. Uranium and plutonium oxide powders and magnetic needles fill up to 70-90% of chamber volume. Ratio of total mass of uranium and plutonium oxide powders to that of magnetic needles is set between 0.30 and 0.65, mainly between 0.40 and 0.50% Then chamber filled with powders and magnetic needles is subjected to deep chilling and powders are mixed up. In the process powders are finely dispersed, loose mass is uniformly mixed up, and fluidity of molding powder is increased. EFFECT: enhanced process efficiency, improved nuclear safety for personnel engaged in mixing up powders. 6 cl, 2 dwg, 1 tbl



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Изобретение относится к области атомной техники и может быть использовано для получения гомогенного ядерного топлива из смеси оксидов урана и плутония с содержанием плутония от 1 до 40 мас.% для ядерных реакторов на быстрых и тепловых нейтронах (МОХ-топлива). Изобретение может быть использовано в производстве таблеток тепловыделяющих элементов активных зон АЭС.

Технология изготовления таблеток оксидного ядерного топлива состоит из операций, связанных с подготовкой пресс-порошка, операции прессования таблеток и операции спекания таблеток в восстановительной атмосфере.

При изготовлении таблеток топлива из нескольких компонентов, например из оксидов урана и плутония, подготовка пресс-порошка включает операцию смешивания компонентов. От этой операции зависят многие характеристики готовой таблетки и в первую очередь гомогенность твердого раствора $(U,Pu)O_2$, плотность, величина зерна, микроструктура.

Известен способ получения таблеток МОХ-топлива, состоящий из подготовки пресс-порошка путем смешивания оксидов урана и плутония, прессования порошка и спекания полученных таблеток, при котором с целью более равномерного распределения компонентов в пресс-порошке проводят измельчение и смешивание компонентов в шаровой мельнице /Мохова В.А. Промышленное производство и опыт эксплуатации (U, $Pu)O_2$ - топлива в реакторах LWRM. ЦНИИАтоминформ. - 1991, выпуск 20, с.26 и 27/.

Известен также способ изготовления таблеток для твэлов реакторов на тепловых нейтронах из (U, $Pu)O_2$, включающий предварительное перемешивание порошков оксидов урана и плутония в V-образном смесителе и размол смеси в течение 20 часов в шаровой или молотковой мельнице с последующими операциями прессования полученной смеси и спекания таблеток /Решетников Ф.Г., Бибилашвили Ю.К. и др. Разработка, производство и эксплуатация тепловыделяющих элементов энергетических реакторов. В 2 кн. Кн. 1. -М.: Энергоатомиздат, 1995, с. 110/.

Однако с помощью этих способов не удается достичь равномерного распределения компонентов в пресс-порошке, и в структуре спеченных таблеток наблюдается наличие двух фаз. Такие таблетки не растворяются полностью в азотной кислоте, что осложняет реализацию замкнутого цикла.

Наиболее близким техническим решением к заявленному способу является способ получения гомогенного ядерного топлива из смеси диоксидов урана и плутония для изготовления таблеток, включающий подготовку пресс-порошка путем смешивания компонентов в вихревом слое, прессование и спекание таблеток /патент RU 2122247, МКИ ^{6} G 21 C 21/00 - прототип/. Вихревой слой создается за счет хаотического перемещения магнитных игл в переменном магнитном поле, которые, увлекая с собой частицы порошков оксидов, одновременно не только перемешивают порошки, но и измельчают их, уплотняют и активируют поверхность частиц порошка. Смешивание порошков производят в рабочем объеме смесителя цилиндрической формы, в который помещают магнитные иглы. Рабочий объем смесителя заполняют на 50 - 70% смесью диоксидов урана и плутония.

Известный способ имеет следующие недостатки. Узкая рабочая зона смесителя (зона переменного магнитного поля), что вызывает необходимость в процессе перемешивания производить возвратно-поступательные движения рабочего объема, представляющего собой герметично закрытый цилиндр, что в свою очередь может привести к недостаточной гомогенности перемешивания компонентов. Цилиндрическая форма рабочего объема накладывает ограничения на загрузку компонентов с точки зрения ядерной безопасности осуществления известного способа перемешивания. Пресс-порошки, получаемые по данному способу, имеют недостаточную насыпную плотность и не обладают текучестью, необходимой для того, чтобы применять его в автоматизированном производстве МОХ-топлива, что вызывает необходимость ввода операции грануляции пресс-порошка на стадии прессования таблеток, понижая производительность изготовления МОХ-топлива и увеличивая энергозатраты. Кроме того, использование известного способа может привести к повышенному загрязнению смеси порошков примесями, например железом до 1000 ppm, образующимся за счет натирания магнитных игл.

Основной технической задачей настоящего изобретения является повышение дисперсности, насыпной массы, текучести получаемой смеси порошков и равномерности перемешивания компонентов по всему объему порошка, повышение производительности процесса и обеспечение ядерной безопасности при смешивании.

Поставленная задача достигается тем, что согласно способу включающему загрузку в камеру смесителя порошков оксидов урана и плутония и магнитных игл, смешивание порошков оксидов урана и плутония с помощью магнитных игл, перемещающихся в камере под воздействием переменного магнитного поля, прессование смеси порошков в таблетку и спекание таблеток, камеру заполняют на 70% - 90% ее объема порошками оксидов урана, плутония и магнитными иглами, при этом отношение суммарной массы порошков оксидов урана и плутония к массе магнитных игл задают от 0,30 до 0,65, преимущественно от 0,40 до 0,50, затем камеру вместе с порошками и магнитными иглами подвергают глубокому охлаждению и проводят смешивание порошков.

Поставленная задача достигается также тем, что в частных вариантах выполнения способа смешивание производят до достижения насыпной плотности от 2,2 г/см^3 до 2,6 г/см^3 в течение 2-5 минут, охлаждение камеры с порошками оксидов урана и плутония и магнитными иглами осуществляют жидким азотом, магнитные иглы изготовляют из материала с твердостью по шкале Роквелла от 69 до 71 ед, коэрцитивной силой от 60 до 770 эрстед,

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например из стали 38ХМЮА, или стали 7ХГ2ВМ, или стали ОХ14АГ12, или стали ЮНДК35Т5, а внутренняя поверхность камеры смесителя имеет в поперечном и продольном сечениях форму овала.

На фиг. 1 приведена общая схема устройства для смешивания порошков по предлагаемому способу.

На фиг.2 приведена микроструктура таблеток, полученных в соответствии с прототипом (а) и предлагаемым способом (б).

Способ осуществляют следующим образом.

Обрабатываемый материал - порошки оксидов урана и плутония - помещают в камеру смесителя, внутренний объем которой имеет в поперечном и продольном сечениях форму овала (фиг. 1), изготовленную из немагнитного материала, например из титана. Туда же засыпают магнитные иглы. Затем камеру смесителя с магнитными иглами и порошками оксидов урана и плутония на несколько минут погружают в хладагент, например жидкий азот, и сразу после этого помещают между электромагнитными индукторами. При подаче электропитания на индукторы в камере смесителя возникает сложное результирующее магнитное поле, приводящее в интенсивное движение магнитные иглы, посредством которых и происходит обработка исходного вещества.

Исходным материалом для обработки по предлагаемому способу была выбрана смесь порошков $U0_2$ и PuO_2 в соотношении 75% : 25% по массе в количестве 5,3 кг. Насыпная плотность смеси была равна 2 г/см^3. Масса магнитных игл, изготовленных из стали 38ХМЮА, составила 10,6 кг. При этом соотношение массы смешиваемых порошков к масое магнитных игл составило 0,5. Эти количества смешиваемых порошков и магнитных игл были рассчитаны заранее для того, чтобы камера смесителя, имеющая объем 4,5 л, была заполнена на 90%. Порошки оксидов урана и плутония с магнитными иглами засыпали в камеру, герметизировали и погружали в жидкий азот. После охлаждения в жидком азоте в течение 5 минут камеру со смесью порошков и магнитных игл помещали в смеситель и проводили операцию перемешивания, измельчения и уплотнения пресс-порошка в течение 5 минут. Затем камеру извлекали из смесителя, охлаждали в течение 20 минут, открывали и отделяли полученную смесь порошков от игл с помощью сит, после чего определяли насыпную плотность и текучесть полученного пресс-порошка. Они составили соответственно 2,4 г/см^3 и 8,0 г/с. Для определения содержания плутония и равномерности его распределения в пресс-порошке случайным образом было отобрано 5 проб по 0,2 г из разных мест объема порошка и исследовано методом кулонометрии с контролируемым потенциалом. По результатам пяти измерений среднее значение содержания Pu в порошке составило 25,00 $\pm$ 0,05%.

Из полученного пресс-порошка на гидравлическом прессе прессовали таблетки при давлении 3 т/см^2, которые затем спекали в шахтной вакуумной электропечи сопротивления СШВЭ - 1.2,5/25 И3, в которой предусмотрен продув газа.

Спекание таблеток проводили в протоке аргоноводородной смеси с содержанием водорода - 7% об. по следующему температурному режиму:

- нагрев со скоростью 600°С/ч до 1750°С;
- выдержка при 1750°С $\pm$ 5°С в течение двух часов;
- охлаждение со скоростью 600°С/ч до комнатной температуры;
- выгрузка спеченных таблеток.

Спеченные таблетки исследовали с помощью металлографического анализа.

Экспериментально получен ряд оптимальных параметров способа, позволяющих повысить равномерность перемешивания, измельчение, насыпную массу, текучесть пресс-порошка и производительность процесса.

Часть магнитных игл под действием бегущей составляющей магнитного поля, перемещается в сторону его движения и постепенно скапливается в нижнем углу камеры штатного смесителя, имеющей форму прямоугольного параллелепипеда. Это приводит к неравномерному распределению магнитных игл по объему рабочей камеры и снижению производительности перемешивания. С целью исключения данного явления внутренний объем камеры смесителя для осуществления заявляемого способа имеет в поперечном и продольном сечениях форму овала (фиг.1), за счет чего магнитные иглы в процессе перемешивания беспрепятственно увлекаются магнитным полем во всем объеме камеры смесителя. Кроме того, выбранная форма камеры смесителя удовлетворяет требованиям ядерной безопасности, что необходимо при работе с МОХ-топливом.

Экспериментально были установлены условия минимального натирания железа и увеличения производительности процесса перемешивания и измельчения порошка.

В результате материалом для изготовления магнитных игл были выбраны сплавы 38ХМЮА, 7ХГ2ВМ, ОХ14АГ12 и ЮНДК35Т5. Эти сплавы характеризуются высокой коэрцетивной силой и остаточной индукцией и соответственно высокой магнитной энергией. Коэрцетивная сила сплавов в 10-20 раз выше, чем у заэвтектоидной стали ШХ15. Кроме того, выбранные сплавы обладают повышенной твердостью R_c=69$\div$71 ед.

Производительность прессования топливных таблеток зависит от насыпной плотности и текучести пресс-порошка. Были определены параметры загрузки обрабатываемого материала и магнитных игл в камеру смесителя для повышения насыпной плотности и текучести пресс-порошков. Установлено, что уплотнение материала для увеличения насыпной плотности и текучести порошков происходит при соотношениях массы порошка к массе магнитных игл, находящемся в интервале от 0,30 до 0,65, и заполнении камеры смесителя на 70% - 90% по объему.

Характеристики пресс-порошков, полученных по способу прототипа и заявляемому способу, и спеченных таблеток, изготовленных из них, в сравнении приведены в таблице и на фиг.2.

Из таблицы и фиг.2 видно, что предлагаемый способ позволяет получать мелкодисперсные, плотные пресс-порошки,

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обладающие высокой текучестью, из которых изготовляют бездефектные таблетки с гомогенным распределением плутония (фиг.2) за счет повышенной активности порошка при спекании.

Использование предлагаемого способа получения гомогенного ядерного топлива обеспечивает по сравнению со способом прототипа новый технический результат:

1. Позволяет получать пресс-порошки с высокой текучестью и насыпной плотностью, что повышает производительность прессования таблеток МОХ-топлива для реакторов на тепловых и быстрых нейтронах.

2. Позволяет получать пресс-порошки с равномерно распределенными по всему объему мелкодисперсными частицами компонентов, что позволяет изготовлять таблетки МОХ-топлива с гомогенным распределением плутония на микроуровне.

3. Повышает производительность приготовления пресс-порошков МОХ-топлива.

4. Обеспечивается выполнение требований по ядерной безопасности при работе с радиоактивными материалами.

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Формула изобретения:

1. Способ получения уран-плутониевого ядерного топлива, включающий загрузку в камеру смесителя порошков оксидов урана и плутония и магнитных игл, смешивание порошков оксидов урана и плутония с помощью магнитных игл, перемещающихся в камере под воздействием переменного магнитного поля, прессование смеси порошков в таблетку и спекание таблеток, отличающийся тем, что камеру заполняют на 70 - 90% ее объема порошками оксидов урана, плутония и магнитными иглами, при этом отношение суммарной массы порошков оксидов урана и плутония к массе магнитных игл задают от 0,30 до 0,65, преимущественно от 0,40 до 0,50, затем камеру вместе с порошками и магнитными иглами подвергают глубокому охлаждению и проводят смешивание порошков.

2. Способ по п. 1, отличающийся тем, что смешивание производят до достижения насыпной плотности смеси от 2,2 до 2,6 г/см^3.

3. Способ по п. 1, отличающийся тем, что смешивание производят в течение 2-5 мин.

4. Способ по п. 1, отличающийся тем, что охлаждение камеры с порошками оксидов урана и плутония и магнитными иглами осуществляют жидким азотом.

5. Способ по п. 1, отличающийся тем, что магнитные иглы изготовляют из материала с твердостью по шкале Роквелла от 69 до 71 ед. и коэрцитивной силой от 60 до 770 эрстед.

6. Способ по п. 5, отличающийся тем, что магнитные иглы изготовляют из стали 38ХМЮА, или стали 7ХГ2ВМ, или стали ОХ14АГ12, или стали ЮНДК35Т5.

7. Способ по п. 1, отличающийся тем, что внутренняя поверхность камеры смесителя имеет в поперечном и продольном сечениях форму овала.

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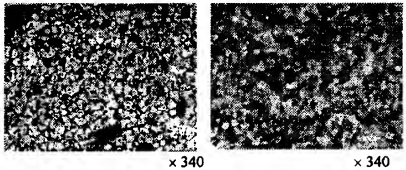
Таблица

Способ обработки порошка	Насыпная плотность	Текучесть	Средний размер частиц	Производи – тельность
	г/см^3	г/с	мкм	кг/ч
Исходный порошок	2,0	Не течёт	< 50	
Прототип	2,1	Не течёт	< 30	6,0
Предлагаемый способ	2,5	8,0	< 20	9,0



× 340 × 340

а б

Фиг. 2

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METHOD FOR PRODUCING HOMOGENEOUS NUCLEAR FUEL

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Applicant(s): SII VSEROSSIJSKIJ NII; G NTS ROSSIJSKOJ FEDERAT; UT NEORGANICHESKIKH MATERIALOV; VARA +
Classification:
- **international:** ***G21C3/62;*** **G21C3/42;** (IPC1-7): G21C3/62
- **European:**
Application number: RU20000111676 20000510
Priority number(s): RU20000111676 20000510

Abstract of **RU 2183035 (C2)**

atomic engineering; nuclear fuel production from mixture of uranium and plutonium oxides. SUBSTANCE: method includes charging the chamber with uranium and plutonium oxide powders and magnetic needles, mixing up these powders by means of magnetic needles, compressing mixture obtained to form pellets, and sintering the pellets. Magnetic needles are moving within chamber under action of variable magnetic field. Uranium and plutonium oxide powders and magnetic needles fill up to 70-90% of chamber volume. Ratio of total mass of uranium and plutonium oxide powders to that of magnetic needles is set between 0.30 and 0.65, mainly between 0.40 and 0.50% Then chamber filled with powders and magnetic needles is subjected to deep chilling and powders are mixed up. In the process powders are finely dispersed, loose mass is uniformly mixed up, and fluidity of molding powder is increased. EFFECT: enhanced process efficiency, improved nuclear safety for personnel engaged in mixing up powders. 6 cl, 2 dwg, 1 tbl

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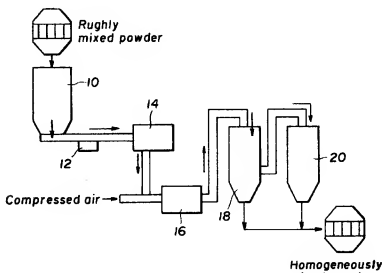
(54) **A method for homogeneously mixing a uranium/plutonium mixed oxide**

(57) A method for homogeneously mixing a uranium/plutonium mixed oxide which is used for the preparation of a uranium/ plutonium mixed oxide fuel. The method comprises weighing a uranium oxide powder, a plutonium oxide powder, and a dry recovered powder prepared by grinding a uranium/plutonium mixed oxide sinter so as to give a predetermined plutonium enrichment; roughly mixing these powders together by means of a mixer; pulverizing and homogeneously mixing the roughly mixed powder by means of a jet mill (16); discharging the homogeneously mixed powder together with compressed air from the jet mill; and separating the mixed powder from the air by means of a first-stage cyclone (18) to recover at least 90% of the discharged powder.

FIG. 2



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Description

[0001] The present invention relates to a method for homogeneously mixing a uranium/plutonium mixed oxide powder, and more particularly to a method wherein a plurality of powders having different densities (specific gravities), such as a uranium oxide powder, a plutonium oxide powder, and a dry recovered powder, are homogeneously mixed together by means of a jet mill and are withdrawn in a homogeneously mixed state. This technique is useful for the preparation of uranium/plutonium mixed oxide pellets.

[0002] The preparation of a uranium/plutonium mixed oxide (MOX) fuel involves the steps of weighing predetermined amounts of a uranium oxide (UO_2) powder, a plutonium oxide (PuO_2) powder, a dry recovered powder prepared by grinding a uranium/plutonium mixed oxide sinter (a powder prepared by grinding sintered uranium/plutonium mixed oxide pellets having, for example, defective appearances, i.e., scrap pellets), and the like so as to give a predetermined plutonium enrichment (percentage addition) and homogeneously mixing the powders. A ball mill or an attritor mill has hitherto been used in the step of homogeneously mixing the uranium/plutonium mixed oxide powder.

[0003] In the ball mill, the feed and recovery of the powders are carried out in such a manner that the powders in a vessel are poured as such into the mill and, after the mixing, the homogeneous powder mixture in the mill is recovered in the vessel by tilting the mill. This method is advantageous in that the powders can be satisfactorily homogeneously mixed together and that the operating conditions (number of revolutions and time) can be readily set, so that it has hitherto been extensively used in the art. The method, however, suffers from low mixing efficiency and the necessity for mixing for a long period of time. Further, the size of the apparatus has been increased for the throughput. In particular, in the preparation of a plutonium-containing nuclear fuel material, the apparatus should be installed within a glove box. Therefore, the apparatus is restricted by the size of the glove box and the consideration of maintenance, so that a large-sized ball mill cannot be installed. An additional problem involved in the ball mill is that the treatment should be carried out batch-wise.

[0004] On the contrary, in the case of the attritor mill, the feed and recovery of the powders are carried out in such a manner that the powders are fed into the mill while vibrating the powders by means of a vibration feeder and that the resultant homogeneous powder mixture is discharged from the mill through piping. In this case, the treatment can be carried out continuously, is suitable for treatment of large amounts of powders, and can be carried out with high mixing efficiency. As with the ball mill, however, the attritor mill has a rotating section driven by a motor and hence is poor in maintenability. Further, since heat of friction is generated during mixing, a cooling mechanism should be provided in order to prevent oxidation of the uranium/plutonium mixed oxide powder. This poses an additional problem that the size of the apparatus should be further increased. Furthermore, due to the construction of the mill, the powders are likely to stay within the mill, leading to an increased exposure dose.

[0006] As well known in the art, jet milling is a method for pulverizing a powder. A jet mill is an apparatus wherein particles are accelerated with the aid of a high-speed gas stream to allow the particles to collide with one another to conduct pulverization. The jet mill is advantageous in that continuous treatment and mass treatment are possible, that the generated heat of friction can be immediately removed, that the size of the device can be reduced, and that the maintenability is good. For these reasons, the jet mill has been used, for example, for pulverizing uranium/plutonium mixed oxide pellets having defective appearances (scrap pellets) to prepare a dry recovered powder. The jet mill, however, has not been used for homogeneously mixing a uranium/plutonium mixed oxide powder.

[0006] The jet mill is equipment which has been originally intended to be used for the pulverization of a ceramic powder but has not been extensively used for mixing purposes. This is because although the jet mill has the function of mixing the powder, a possible range of mixing is limited. The reason for this is as follows. In the jet mill, the powders are fed and discharged in a manner utterly different from that in the case of the above-described ball mill and the like. Specifically, the powders are fed into the jet mill with the aid of compressed air and, after the pulverization, the mixture of the pulverized powder with the air stream is discharged from the jet mill and then separated into the powder (solid) and the gas by means of a cyclone, a bag filter or the like, followed by the recovery of the separated powder only in a vessel.

[0007] In the above-described method for feeding and discharging powders using the jet mill, when powders having different compositions with different densities are mixed, unfavorably the dissimilar powders thus mixed are separated again into one another due to their difference in density in the course of the separation of the solid from the gas after the discharge. This leads to a variation in the composition of the resultant powder mixture. Therefore, the contemplated homogeneous mixing cannot be achieved. For this reason, the jet mill has been used in most cases in pulverization of ceramic powders having one and the same composition (for example, a powder prepared by crushing a sinter forming a solid solution on an atomic level) and the like, but has not been used in applications where dissimilar powders having different densities are homogeneously mixed together (mixing while pulverizing).

[0008] As described above, in the preparation of a uranium/plutonium mixed oxide fuel, a uranium oxide powder, a plutonium oxide powder, a dry recovered powder and the like should be homogeneously mixed

together while pulverizing. If the homogenization-mixing is unsatisfactory, a portion rich in the plutonium component, called a "plutonium spot", is created within the sintered pellet. The presence of such a plutonium spot causes this portion to intensively undergo fission during exposure of the pellet, creating a high-temperature hot spot. The plutonium spot present within the pellet has no significant influence. On the other hand, when the plutonium spot is present around the surface of the pellet, that portion becomes hot, greatly affecting a metallic cladding tube. In particular, the rise of the spot temperature sometimes causes the cladding tube to be melted, leading to a serious trouble, that is, fuel failure. For this reason, the size of the plutonium spot and the plutonium concentration of the pellet are strictly restricted, so that preferably the plutonium concentration should be as uniform as possible and the diameter of the plutonium spot should be as small as possible.

[0009] Various powders used in the preparation of a uranium/plutonium mixed oxide fuel are significantly different from one another in powder density. Specifically, the density of the plutonium oxide powder, the lowest-density powder, is about 2 g/cc, whereas the density of the dry recovered powder, the highest-density powder, is about 6 g/cc, that is, three times larger than that of the lowest-density powder. For this reason, when the jet mill is used, although the homogenization-mixing per se in the jet mill can be successfully carried out without posing any problem, the powders are again separated into one another due to their density difference in the course of the separation of the powder mixture from the gas after the discharge from the mill. Therefore, a plutonium-rich portion and a plutonium-lean portion are created in the powder, so that the contemplated satisfactory homogenization cannot be achieved.

[0010] According to the present invention, there is provided a method for homogeneously mixing a uranium/ plutonium mixed oxide comprising: weighing a uranium oxide powder, a plutonium oxide powder, and a dry recovered powder prepared by grinding a uranium/plutonium mixed oxide sinter so as to give a predetermined plutonium enrichment; roughly mixing these powders together by means of a mixer; pulverizing and homogeneously mixing the roughly mixed powder by means of a jet mill; discharging the homogeneously mixed powder together with compressed air from the jet mill; and separating the mixed powder from the air by means of a first-stage cyclone to recover at least 90% of the discharged powder.

[0011] The reason why at least 90% of the discharged powder is recovered at once by means of a first-stage cyclone is that, even when there is about three times as great a difference in density, the powders are less likely to be re-separated from one another in the course of recovery. The recovery of at least 90% can be realized by regulating the amount of the compressed air fed into the jet mill (by increasing the amount of the air to increase the speed of the air stream). Alternatively, this can be attained also by varying the configuration of the first-stage cyclone, for example, by reducing the inner diameter of the cyclone or by increasing the length of the cyclone.

[0012] An example of the present invention will now be described in detail with reference to the accompanying drawings, in which:

Fig. 1 is a process diagram for the preparation of uranium/plutonium mixed oxide fuel pellets;
Fig. 2 is a diagram illustrating a homogenization-mixing process using a jet mill; and,
Fig. 3 is a diagram showing the structure of one example of the jet mill.

[0013] The preparation of uranium/plutonium mixed oxide fuel pellets is carried out according to a preparation process as shown in Fig. 1. At the outset, a uranium oxide powder, a plutonium oxide powder, and a dry recovered powder (a powder prepared by grinding scrap pellets) are weighed so as to give a predetermined plutonium enrichment. Next, these powders are roughly mixed together by means of a mixer (though this step is unnecessary in the conventional method wherein a ball mill is used in the homogenization-mixing). In the stage of rough mixing, the powders having respective compositions are merely mixed together without any change in the size of the particles. If pellets are prepared from this roughly mixed powder, the plutonium oxide particle portion, even by sintering, does not satisfactorily yield a solid solution with uranium oxide, causing the plutonium oxide component to be left as a dense portion (a plutonium spot). In order to minimize the plutonium spot and to uniform the plutonium concentration, it is necessary to finely grind the particles of each powder and to homogeneously mix the fine powders. This step is called "homogenization-mixing." The powder is granulated after the homogenization-mixing for facilitating the molding, and the resultant granules are then molded into a desired shape and sintered to dissolve plutonium and uranium in each other to form a solid solution. After inspection, the product serves as a uranium/plutonium mixed oxide fuel pellet.

[0014] The step of homogenization-mixing by means of a jet mill according to the present invention is carried out by using a system as shown in Fig. 2. The roughly mixed powder is transferred to a storage tank 10 and then fed by means of a feeder 12 into a constant rate feeder 14. The roughly mixed powder is then transferred from the feeder 14 at a constant feed rate, accelerated by compressed air, and fed into a jet mill 16, where pulverization and mixing (homogenization-mixing) are carried out. The homogenized and mixed powder, together with the compressed air stream, is discharged from the jet mill 16, and the powder is separated from the air through a cyclone 18 and a cyclone 20 with a built-in bag filter. The powder is collected in a storage tank provided at the lower parts of both the cyclone 18 and the

cyclone 20 with a built-in bag filter and recovered as a homogeneously mixed powder. This homogeneously mixed powder is then transferred to the following step of granulation.

[0015] In the jet mill, particles accelerated to approach the velocity of sound by means of compressed air of 6 to 7 atm are allowed to collide with one another within the mill to pulverize the powder particles by utilizing the impact of the collision. In this case, mixing of dissimilar powders is simultaneously carried out. Fig. 3 shows an example of a suitable jet mill. The jet mills have various types of sizes and the one shown in Fig. 3 is of a vertical type. Besides this, there is a horizontal type jet mill like a particle accelerator. The roughly mixed powder is fed through a powder inlet 30, while the compressed air is fed through a compressed air inlet 32. The powder is accelerated by the compressed air, further accelerated by a Venturi nozzle 34, and fed into a mill body 36. Compressed air passed through a grinding nozzle 38 is blown into the mill body through the wall of the mill to permit the powder particles to collide with one another at violent speed near the velocity of sound in a mixing-pulverization zone 40. Thus, the particles are pulverized and at the same time are mixed together. The resultant pulverized powder is separated in a classification zone 42 by a centrifugal force into coarse particles and fine powder. The fine powder is discharged through an output 44 outside the mill, while the coarse particles are returned to the mixing-pulverization zone 40. Thus, pulverization and mixing are carried out.

[0016] In the homogenization-mixing system shown in Fig. 2, the powders constituting the roughly mixed powder to be fed into the jet mill 16 are significantly different from one another in powder density. Specifically, the density of the plutonium oxide powder, the lowest-density powder, is about 2 g/cc, whereas the maximum density of the dry recovered powder, the highest-density powder, is about 6 g/cc, that is, three times larger than that of the lowest-density powder. For this reason, despite the homogenization-mixing within the body of the jet mill 16, there is a possibility that the powders constituting the homogeneously mixed powder are again separated from one another due to their density difference in the course of the separation of the powder from the gas after being discharged from the jet mill. In the cyclone 18, a powder having a relatively high density and a powder having a relatively large particle diameter are separated and collected in the lower part of the cyclone 18, while, in the cyclone 20 with a built-in bag filter, a powder having a relatively low density and a powder having a relatively small particle diameter are separated and collected by the lower part of the cyclone 20. When the powder collected by the cyclone 18 and the powder collected by the cyclone 20 are recovered in the same vessel, the mixed powder has a plutonium-rich portion and a plutonium-lean portion, so that no desired results can be obtained.

[0017] In order to evade this situation in the present invention, the recovery of the powder in the first-stage cyclone 18 is regulated to be at least 90%. The regulation of the recovery can be realized by regulating the amount of the compressed air fed into the jet mill (by increasing the amount of the air to increase the speed of the air stream). The practical numerical value of the amount of the compressed air fed into the jet mill necessary for this end can be experimentally determined although it varies depending upon the size and structure of the equipment used. Besides this, varying the configuration of the first-stage cyclone, for example, reducing the inner diameter of the cyclone or increasing the length of the cyclone, also enables the recovery of at least 90% of the discharged powder in the first-stage cyclone. The reason why at least 90% of the discharged powder should be recovered in the first-stage cyclone is that this value has been experimentally found to be effective in preventing the re-separation of the powders constituting the uranium/plutonium mixed oxide powder and having three times as great a difference in density.

[0018] The α autoradiograph of a pellet prepared by the homogenization-mixing in a jet mill according to the present invention was compared with that of a pellet prepared by the homogenization-mixing in a ball mill according to the prior art method. The "α autoradiograph" is a photograph prepared by pressing the pellet against cellulose and then conducting exposure and development. Since a portion that is richer in α-rays (that is, a portion that is richer in plutonium) destroys the cellulose structure more severely and looks black in the photograph, the diameter and amount of the plutonium spot can be observed. According to this method, in the case of the pellet prepared by using a roughly mixed powder, a large number of large black spots (plutonium spots) are observed, and the size of each spot is up to about 0.3 mm. This state does not satisfy the requirements for the plutonium spot. By contrast, according to the method of the present invention, the plutonium spot is hardly observed and, if any, has a size on the order of about several μm, confirming that the method of the present invention using a jet mill can realize homogenization-mixing to an extent comparable to that attained by the ball mill.

[0019] The method of the present invention is applied to the homogenization-mixing of a uranium/plutonium mixed oxide powder. In addition, the method can be applied also to homogenization-mixing of a uranium/gadolinia (Gd_2O_3) mixed oxide powder or a uranium/plutonium/gadolinia mixed oxide powder.

[0020] As being understood from the foregoing, according to the method for homogeneously mixing a uranium/plutonium mixed oxide by means of a jet mill, the treatment is carried out continuously, thus enabling treatment of the powders in a large amount, and hardly creates an adverse effect of the heat of friction during mixing (although this heat of friction is generated to some extent, the generated heat is immediately removed because mixing is carried out in a large amount

of an air stream), so that the powder is hardly oxidized. Further, regarding the system, the size of the apparatus can be reduced and the maintenance can be easily carried out, so that the apparatus can be easily housed in a glove box. Further, since there is no rotating section driven by a motor or the like, no deposition of powders on the rotating section occurs and the frequency of failures created by friction is small. Furthermore, the powders hardly stay within the mill and the exposure dose is not increased, while there is no need to provide a cooler. Furthermore, the homogeneity of the resultant mixed powder is comparable to that of the mixed powder prepared by homogenization-mixing using a ball mill.

Claims

1. A method for homogeneously mixing a uranium/plutonium mixed oxide comprising the steps of:

   passing a mixture of a uranium oxide powder, a plutonium oxide powder, and a dry recovered powder prepared by grinding a uranium/plutonium mixed oxide sinter so as to give a predetermined plutonium enrichment through a jet mill to pulverize and homogeneously mix the powders; discharging the homogeneously mixed powder in a stream of compressed air from the jet mill; and,
   recovering at least 90% of the homogeneously mixed powder when passing the stream of powder and compressed air through a first stage cyclone.

2. A method according to claim 1, in which regulation of the recovery of the homogeneously mixed powder is achieved by controlling the amount of compressed air introduced into the jet mill.

3. A method according to claim 1 or 2, further comprising the step of premixing the powders together before feeding them into the jet mill.

4. A method for homogeneously mixing a uranium/plutonium mixed oxide comprising:

   weighing a uranium oxide powder, a plutonium oxide powder, and a dry recovered powder prepared by grinding a uranium/plutonium mixed oxide sinter so as to give a predetermined plutonium enrichment;
   roughly mixing these powders together by means of a mixer; pulverizing and homogeneously mixing the roughly mixed powder by means of a jet mill;
   discharging the homogeneously mixed powder together with compressed air from the jet mill; and,
   separating the mixed powder from the air by means of a first-stage cyclone to cover at least 90% of the discharged powder.

FIG. 1

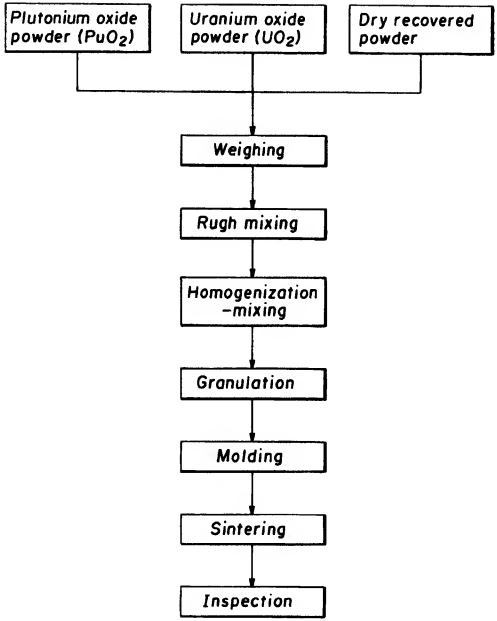

Plutonium oxide powder (PuO_2)
Uranium oxide powder (UO_2)
Dry recovered powder
Weighing
Rugh mixing
Homogenization -mixing
Granulation
Molding
Sintering
Inspection

FIG. 2

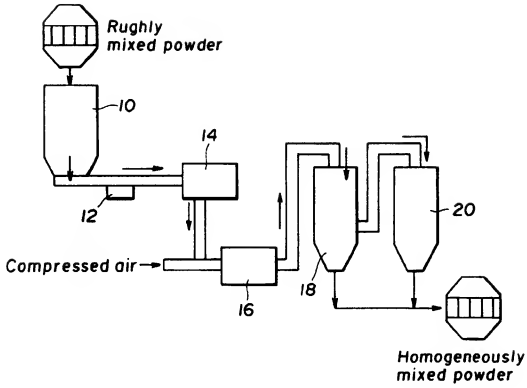
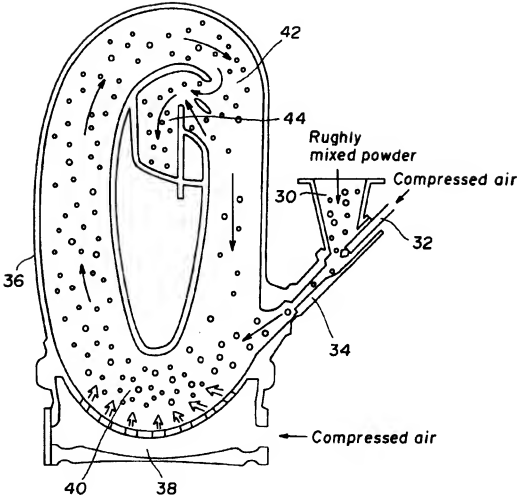

Rughly
mixed powder
10
14
12
20
Compressed air
16
18
Homogeneously
mixed powder

FIG. 3


42
44
Rughly
mixed powder
Compressed air
30
32
36
34
Compressed air
40
38